

Time dependence of photon correlations in a three-level atomic cascade

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The question of the temporal correlation of photons arising from a three-level atomic cascade is considered by calculating the joint probability of photoelectric detection for two detectors placed in the far field of the atomic emission. The analysis is carried out in the Heisenberg picture and involves the use of dipole, adiabatic (Weisskopf-Wigner), and rotating-wave approximations. Except for a small interval about zero time delay, which is however too short to be detected, it is found that the temporal correlation between emitted photons is one of sequential but otherwise statistically independent events, even when Landau's generalized decay and frequency-shift terms are included. This result is compared with a well-known expression for the joint spectral density of the emitted photons.

I. INTRODUCTION

The measurement of the correlation of photons emitted in an atomic cascade is an accepted technique for determining lifetimes of excited atomic levels.¹⁻⁴ For an arrangement of levels such as shown in Fig. 1, with level 3 initially excited, the lifetime of level 2 is found by recording the distribution of time delays between an initial detection arising from the $3 \rightarrow 2$ transition and a subsequent detection from the $2 \rightarrow 1$ decay. Implicit in this scheme is the assumption that the emission of the first photon projects the atom into level 2 with absolute certainty and that there are no residual correlations between successive photons. While these assumptions are certainly plausible, there appears to be in the literature no corresponding calculation of the actual joint probability for photoelectric detection.

Another important application of the measurement of photon correlations is found in tests of Bell inequalities in studies of polarization correlations for photons emitted in an atomic cascade. Extensive reviews of this work are available.⁵⁻⁷ Generally one is interested in the coincidence rate as a function of the relative polarization of emitted pairs of photons from a cascade. The actual time dependence of the joint detection probability may be recorded, or more simply the number of coincidences may be obtained by employing a relatively wide time window triggered by the initial event. In the former case the joint detection probability as a function of time delay between events is well described by a decaying exponential of time constant given by the lifetime of the intermediate state.⁸ In either case quantum-mechanical predictions violating Bell inequalities do not depend upon the dynamics of the photon emissions but rather upon consideration of the angular momenta of initial and final states, and on the prescription chosen for extracting probabilities from a two-particle state.

Treatments that do yield dynamical information about

the intensity correlation of the field emitted in a cascade have been given by Loudon⁹ and by Aspect *et al.*^{6(b)} For the case of continuous but incoherent excitation of level 3, Loudon⁹ uses rate equations together with the quantum regression theorem to show that the decay $3 \rightarrow 2 \rightarrow 1$ proceeds in a sequential but otherwise uncorrelated fashion, in agreement with the numerous observations quoted above. Scattering theory has been employed by Aspect *et al.*^{6(b)} to analyze the photon correlations in the excitation and decay of a cascade, including the role of Zeeman levels. Agarwal and Jha¹¹ treat the related problem of resonant Raman scattering.

A different perspective on the question of photon correlations is provided by a calculation of the joint spectral density $J(\omega, \omega')$ from the decay sequence $3 \rightarrow 2 \rightarrow 1$ for an atom initially excited to state 3. Weisskopf and Wigner¹⁰ and others^{11,12} have considered this problem and find

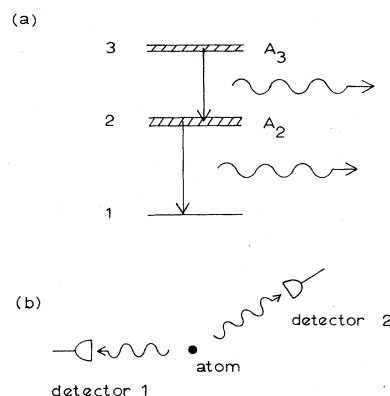


FIG. 1. (a) Level scheme for the atomic cascade $3 \rightarrow 2 \rightarrow 1$. The transition $3 \rightarrow 1$ is forbidden; A_2 and A_3 are the Einstein A coefficients for the levels 3 and 2, respectively. (b) Detectors in the far field of the atomic radiation for registering the joint probability of photoelectric detection at points (\mathbf{r}_1, t_1) and (\mathbf{r}_2, t_2) .

$$J(\omega, \omega') \propto \frac{\omega' \omega}{\omega_{21} \omega_{32}} \left[\frac{\beta_3 / \pi}{\beta_3^2 + (\omega_{31} - \omega - \omega')^2} \right] \\ \times \left[\frac{\beta_2 / \pi}{\beta_2^2 + (\omega_{21} - \omega')^2} \right],$$

where the notation refers to Fig. 1 with $\omega_{32}, \omega_{21}, \omega_{31}$ as the transition frequencies and β_3, β_2 as half the Einstein A coefficients for the respective transitions. As Weisskopf and Wigner point out,¹⁰ the expression for $J(\omega, \omega')$ "shows the statistical dependence of the frequencies ω, ω' of the two successively emitted quanta." This dependence is further illustrated by passing to the limit of a very narrow excited state 3 ($\beta_3 \rightarrow 0$), for which

$$J(\omega, \omega') \rightarrow \frac{\omega' \omega}{\omega_{21} \omega_{32}} \delta(\omega_{31} - \omega - \omega') \left[\frac{\beta_2 / \pi}{\beta_2^2 + (\omega_{21} - \omega')^2} \right].$$

Again from Weisskopf and Wigner, since ω' can vary over the range β_2 , this variation "must be compensated by ω , leading to a statistical coupling of the two frequencies and a broadening of the line ($3 \rightarrow 2$)."

This observation of the statistical coupling of the emission frequencies on the one hand, and the need for explicit quantum calculations of the temporal correlation of emitted photons on the other, have motivated the analysis presented in this paper. For an atom of an arbitrary initial excitation including possible coherence among the atomic states, we wish to determine the probability to detect two photons, one at time t and a second at time $t + \tau$, as the atom decays spontaneously into the vacuum. Given this result for temporal correlations we shall then return to examine the Weisskopf-Wigner result for frequency correlations. The problem of correlations in a sequential decay has also been treated by Henin, both

within and outside the framework of quantum electrodynamics.¹³

The outline of the paper is the following. In Sec. II we recall the work of Glauber¹⁴ which expresses the joint probability of photoelectric detection for an experiment such as that shown in Fig. 1 in terms of an intensity correlation function of the field. In Sec. III we derive equations of motion for the coupled system of atom plus quantized field within the dipole, rotating-wave, and "adiabatic" approximations. In Secs. IV and V these equations are used to compute field correlation functions and probabilities of photoelectric detection for an arbitrary initial atomic state and for the vacuum state of the field. Section VI serves as a summary.

II. INTENSITY CORRELATION RELATED TO PHOTOELECTRIC DETECTION PROBABILITY

The circumstance that we consider is shown in Fig. 1. The levels (1,2,3) are assumed to be nondegenerate with the transitions ($3 \rightarrow 2$) and ($2 \rightarrow 1$) being allowed electric dipole transitions and with the transition ($3 \rightarrow 1$) forbidden. The atom is taken to be in some initial state $|\phi_A\rangle$, and the field state is chosen to be the vacuum state $|\{0\}\rangle$. We will investigate the photon statistics of the field arising from the atomic cascade with photodetectors placed in the far field of the atomic emission. Since in general the two frequencies involved in the cascade will be substantially different, we require a detection probability for the case of polychromatic light. Following the work of Glauber,^{14(b)} we write the joint probability $P_2(x_1; x_2)$ of a photoelectric event at $x_1 \equiv (r_1, t_1)$ within an interval Δt_1 followed by one at $x_2 \equiv (r_2, t_2)$ within an interval Δt_2 as^{15,16}

$$P_2(x_1; x_2) = \int_{t_1}^{t_1 + \Delta t_1} dt'_1 \int_{t_2}^{t_2 + \Delta t_2} dt'_2 \int_{t_2}^{t_2 + \Delta t_2} dt''_2 \int_{t_1}^{t_1 + \Delta t_1} dt''_1 k_{ji}^{(1)}(t'_1 - t''_1) k_{ml}^{(2)}(t'_2 - t''_2) \\ \times \langle \mathcal{T} [\hat{A}_i^{(-)}(r_1, t'_1) \hat{A}_l^{(-)}(r_2, t'_2) \hat{A}_m^{(+)}(r_2, t''_2) \hat{A}_j^{(+)}(r_1, t''_1)] \rangle. \quad (2.1)$$

The detection process is modeled with the $\hat{\mathbf{p}} \cdot \hat{\mathbf{A}}$ interaction, with $\hat{\mathbf{p}}$ as the momentum of an electron in the detector surface and $\hat{\mathbf{A}}$ as the vector potential of the total field radiated by the atom, with positive and negative frequency components $\hat{\mathbf{A}}^{(+)}, \hat{\mathbf{A}}^{(-)}$, respectively. The detection intervals $\Delta t_1, \Delta t_2$ are assumed to be long compared to the optical period, and as a consequence the rotating-wave approximation is made. In (2.1) $k_{ij}(t)$ is the response function of the photodetector (1 or 2), defined as in Ref. 15, and a summation over Cartesian indices (i, j) and (l, m) is implicit. \mathcal{T} is the time-ordering symbol that places positive frequency operators in ascending order of time argument from the right and negative frequency operators in ascending order from the left. The time ordering is essential in (2.1) since in the present problem the field generat-

ed by the atom does not necessarily commute with itself at different space-time points.

As is clear from (2.1) and as was stressed in Ref. 14(b), for polychromatic light the joint probability of photo-detection is not related to the light intensities at the detectors in any simple way. In order to emphasize the essential features of the photon correlations in the atomic cascade, we will consider the case of an "ideal" detector defined by^{14(b)}

$$k_{ij}(t) = k_0 \delta_{ij} \delta(t). \quad (2.2)$$

The reduction of the polarization dependence to a Kronecker delta function is justified in Ref. 15, while the Dirac delta function in time implies an (unrealistic) infinite bandwidth for detection or a zero electronic correla-

tion time.¹⁷ However, we will see in Sec. V that Eq. (2.2) is readily modified to encompass most experimental situations. We note in passing that (2.2) is one of two physically reasonable choices for the definition of an ideal detector. An ideal detector with a uniform quantum efficiency over all frequencies would be defined with the derivative of a Dirac delta function appearing in (2.2).

For the case of identical detectors as defined by (2.2),

$$\Gamma^{(2,2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) \equiv \langle \mathcal{T} [\hat{A}_i^{(-)}(\mathbf{r}_1, t_1) \hat{A}_j^{(-)}(\mathbf{r}_2, t_2) \hat{A}_j^{(+)}(\mathbf{r}_2, t_2) \hat{A}_i^{(+)}(\mathbf{r}_1, t_1)] \rangle. \quad (2.4)$$

Note that $\Gamma^{(2,2)}$ is a normally ordered and time-ordered correlation function. The integrals in (2.3) range over the intervals $\Delta t_1 = \Delta t_2 = \Delta t$ that represent the finite resolution of the detector. The time Δt is taken to be large compared with the intrinsic response time of the detector [strictly zero by (2.2)], as, for example, at the photocathode, and large compared to the optical period. The interval is, however, meant to be small with respect to the lifetimes of the states involved in the cascade. In our discussion of the cascade we will also require the probability for single photoelectric detection, which for the ideal detector specified by (2.2) is^{14,15}

$$P_1(x) = k_0 \int_t^{t+\Delta t} dt' \langle \hat{A}_i^{(-)}(\mathbf{r}, t') \hat{A}_i^{(+)}(\mathbf{r}, t') \rangle. \quad (2.5)$$

Our task then is to obtain equations of motion for atomic and field operators and from these equations to calculate $\Gamma^{(2,2)}(x_1; x_2)$ and hence $P_2(x_1; x_2)$.

III. DERIVATION OF EQUATIONS OF MOTION

To describe the atomic cascade we will employ the Hamiltonian for a one-electron atom interacting with the electromagnetic field. Our analysis is to be carried out in the nonrelativistic case and will closely follow the treatment of spontaneous emission in multilevel systems given by Milonni.¹⁸ We write the Hamiltonian of the system in the form

$$\hat{H} = \hat{H}_{\text{atom}} + \hat{H}_{\text{field}} + \hat{H}_{\text{int}} \quad (3.1)$$

with

$$\hat{H}_{\text{atom}} = \frac{\hat{p}^2}{2m} + V(\mathbf{r}), \quad (3.2)$$

$$\hat{H}_{\text{field}} = \sum_{\mathbf{k}, \lambda} \hbar \omega_{\mathbf{k}} \hat{a}_{\mathbf{k}\lambda}^\dagger \hat{a}_{\mathbf{k}\lambda}, \quad (3.3)$$

and

$$\hat{H}_{\text{int}} = -\frac{e}{mc} \hat{\mathbf{A}}(0, t) \cdot \hat{\mathbf{p}}(t). \quad (3.4)$$

The dipole approximation has been made by evaluating the vector potential $\hat{\mathbf{A}}$ at the position of the atomic nucleus, which we assume to be the origin, and the \hat{A}^2 term has been dropped. The notation is that of Ref. 18 with $\hat{\mathbf{A}}$

we find from (2.1)

$$P_2(x_1; x_2) = k_0^2 \int_{t_1}^{t_1 + \Delta t_1} dt'_1 \int_{t_2}^{t_2 + \Delta t_2} dt'_2 \Gamma^{(2,2)}(\mathbf{r}_1, t'_1; \mathbf{r}_2, t'_2), \quad (2.3)$$

with the fourth-order field correlation function $\Gamma^{(2,2)}$ defined by

evaluated in the Coulomb gauge and expanded in terms of plane waves in a volume V as

$$\hat{\mathbf{A}}(\mathbf{r}, t) = \sum_{\mathbf{k}, \lambda} \left[\frac{2\pi\hbar c^2}{\omega_{\mathbf{k}} V} \right]^{1/2} [\hat{a}_{\mathbf{k}\lambda}(t) e^{i\mathbf{k} \cdot \mathbf{r}} \boldsymbol{\epsilon}_{\mathbf{k}\lambda} + \text{H.c.}]. \quad (3.5)$$

In the Heisenberg picture the photon annihilation and creation operators for the mode of wave vector \mathbf{k} and polarization index $\lambda = 1, 2$ obey the equal-time commutation relation

$$[\hat{a}_{\mathbf{k}, \lambda}(t), \hat{a}_{\mathbf{k}', \lambda'}^\dagger(t)] = \delta_{\mathbf{k}, \mathbf{k}'}^3 \delta_{\lambda, \lambda'}. \quad (3.6)$$

In terms of the eigenstates $|n\rangle$ of the atomic Hamiltonian \hat{H}_a , any atomic operator $\hat{B}(t)$ may be expressed as

$$\hat{B}(t) = \sum_m \sum_n \langle n | \hat{B}(0) | m \rangle \hat{\sigma}_{nm}(t), \quad (3.7)$$

with

$$\hat{\sigma}_{nm}(t) = \exp(i\hat{H}t/\hbar) \hat{\sigma}_{nm}(0) \exp(-i\hat{H}t/\hbar)$$

and $\hat{\sigma}_{nm}(0) = |n\rangle \langle m|$. The atomic operators $\hat{\sigma}_{nm}(t)$ satisfy the commutation relation

$$[\hat{\sigma}_{ij}(t), \hat{\sigma}_{kl}(t)] = \delta_{jk} \hat{\sigma}_{il}(t) - \delta_{il} \hat{\sigma}_{kj}(t), \quad (3.8)$$

and are such that

$$\hat{\sigma}_{ij}(t) \hat{\sigma}_{kl}(t) = \hat{\sigma}_{il}(t) \delta_{jk}. \quad (3.9)$$

The representation of (3.7) allows us to rewrite the Hamiltonian as

$$\begin{aligned} \hat{H} = & \sum_n E_n \hat{\sigma}_{nn}(t) + \sum_{\mathbf{k}, \lambda} \hbar \omega_{\mathbf{k}} \hat{a}_{\mathbf{k}\lambda}^\dagger(t) \hat{a}_{\mathbf{k}\lambda}(t) \\ & + i\hbar \sum_{\mathbf{k}, \lambda} \sum_{n, m} D_{\mathbf{k}\lambda nm} \hat{\sigma}_{nm}(t) [\hat{a}_{\mathbf{k}\lambda}(t) + \hat{a}_{\mathbf{k}\lambda}^\dagger(t)], \end{aligned} \quad (3.10)$$

where

$$D_{\mathbf{k}\lambda nm} \equiv \frac{1}{\hbar} \left[\frac{2\pi\hbar}{\omega_{\mathbf{k}} V} \right]^{1/2} \omega_{mn} \boldsymbol{\mu}_{nm} \cdot \boldsymbol{\epsilon}_{\mathbf{k}\lambda}, \quad (3.11a)$$

$$\hbar \omega_{mn} \equiv E_m - E_n, \quad (3.11b)$$

and

$$\boldsymbol{\mu}_{nm} \equiv \langle n | e\mathbf{r} | m \rangle. \quad (3.11c)$$

E_m is the (unperturbed) energy of the m th atomic state

and μ_{nm} is the transition moment for electric dipole transitions between states n and m . For simplicity μ_{nm} and $\epsilon_{k\lambda}$ will be taken to be real.

The Heisenberg equations of motion follow directly from Eqs. (3.6), (3.8), and (3.10), and take the form

$$\dot{\hat{a}}_{k\lambda}(t) = -i\omega_k \hat{a}_{k\lambda}(t) + \sum_{l,m} D_{k\lambda lm} \hat{\sigma}_{lm}(t), \quad (3.12)$$

$$\begin{aligned} \dot{\hat{\sigma}}_{ij}(t) = & -i\omega_{ji} \hat{\sigma}_{ij}(t) \\ & + \sum_{k,\lambda} \sum_{l,m} D_{k\lambda lm} \{ [\delta_{jl} \hat{\sigma}_{im}(t) - \delta_{im} \hat{\sigma}_{lj}(t)] \hat{a}_{k\lambda}(t) \\ & + \hat{a}_{k\lambda}^\dagger(t) [\delta_{jl} \hat{\sigma}_{im}(t) - \delta_{im} \hat{\sigma}_{lj}(t)] \}, \end{aligned} \quad (3.13)$$

where the second equation has been written in normal order.

Assuming the atom-field interaction to be weak, we make the so-called adiabatic approximation,

$$\hat{\sigma}_{lm}(t_1) \cong \hat{\sigma}_{lm}(t) \exp[-i\omega_{ml}(t_1 - t)], \quad (3.14)$$

which we expect to be valid for times long compared with an optical period but short compared with the atomic lifetime. This approximation has been discussed extensively¹⁹ and its equivalence to the Weisskopf-Wigner approximation demonstrated.²⁰ Formally integrating Eq. (3.12) and introducing Eq. (3.14) leads to

$$\hat{a}_{k\lambda}(t) = \hat{a}_{k\lambda}(0) e^{-i\omega_k t} + \sum_{l,m} D_{k\lambda lm} \int_0^t dt_1 e^{i\omega_k(t_1 - t)} \hat{\sigma}_{lm}(t_1), \quad (3.15a)$$

$$\hat{a}_{k\lambda}(t) \cong \hat{a}_{k\lambda}(0) e^{-i\omega_k t} + \sum_{l,m} D_{k\lambda lm} \hat{\sigma}_{lm}(t) \int_0^t dt_1 e^{i(\omega_k - \omega_{ml})(t_1 - t)}. \quad (3.15b)$$

This equation may be used to eliminate $\hat{a}_{k\lambda}(t)$ in Eq. (3.13), resulting in

$$\begin{aligned} \dot{\hat{\sigma}}_{ij}(t) = & -i\omega_{ji} \hat{\sigma}_{ij}(t) + i \sum_m (\gamma_{jmmj} - \gamma_{immi}) \hat{\sigma}_{ij}(t) - \sum_m (\beta_{jmmj} + \beta_{immi}) \hat{\sigma}_{ij}(t) \\ & + \sum_{k,\lambda} \sum_{l,m} D_{k\lambda lm} \{ [\delta_{jl} \hat{\sigma}_{im}(t) - \delta_{im} \hat{\sigma}_{lj}(t)] \hat{a}_{k\lambda}(0) e^{-i\omega_k t} + \hat{a}_{k\lambda}^\dagger(0) e^{i\omega_k t} [\delta_{jl} \hat{\sigma}_{im}(t) - \delta_{im} \hat{\sigma}_{lj}(t)] \} \\ & - \sum_m \sum_{\substack{p \\ p \neq j}} \Gamma_{jmpp} \hat{\sigma}_{ip}(t) - \sum_m \sum_{\substack{p \\ p \neq i}} \Gamma_{impp}^* \hat{\sigma}_{pj}(t) + \sum_m \sum_p \Gamma_{mijp} \hat{\sigma}_{mp}(t) + \sum_m \sum_p \Gamma_{mjip}^* \hat{\sigma}_{pm}(t), \end{aligned} \quad (3.16)$$

with

$$\begin{aligned} \Gamma_{lmnp} \equiv & \frac{1}{\hbar^2} \sum_{k,\lambda} \frac{2\pi\hbar}{\omega_k V} \omega_{lm} \omega_{pn} (\mu_{lm} \cdot \epsilon_{k\lambda}) (\mu_{np} \cdot \epsilon_{k\lambda}) \\ & \times \int_0^t dt_1 e^{i(\omega_k - \omega_{pn})(t_1 - t)}. \end{aligned} \quad (3.17)$$

In the limit $V \rightarrow \infty$ and for times t long compared to $(\omega_{pn})^{-1}$, we have²⁰

$$\Gamma_{lmnp} = \beta_{lmnp} - i\gamma_{lmnp}, \quad (3.18)$$

where

$$\beta_{lmnp} \equiv \frac{2}{3\hbar c^3} \omega_{lm} (\omega_{pn})^2 (\mu_{lm} \cdot \mu_{np}) U(\omega_{pn}), \quad (3.19)$$

$$\gamma_{lmnp} \equiv \frac{2}{3\pi\hbar c^3} \omega_{lm} \omega_{pn} (\mu_{lm} \cdot \mu_{np}) P \int \frac{\omega d\omega}{\omega - \omega_{pn}}, \quad (3.20)$$

and U is the unit step function.

At this point we wish to restrict our attention from the total atomic Hilbert space to the three-level subspace shown in Fig. 1. Including only the states 1,2,3 of Fig. 1, we obtain from (3.16) the following three-level equations of motion:²¹

$$\dot{\hat{\sigma}}_{11}(t) = \hat{X}_{11}(t) + A_2 \hat{\sigma}_{22}(t), \quad (3.21a)$$

$$\dot{\hat{\sigma}}_{22}(t) = \hat{X}_{22}(t) - A_2 \hat{\sigma}_{22}(t) + A_3 \hat{\sigma}_{33}(t), \quad (3.21b)$$

$$\dot{\hat{\sigma}}_{33}(t) = \hat{X}_{33}(t) - A_3 \hat{\sigma}_{33}(t), \quad (3.21c)$$

$$\begin{aligned} \dot{\hat{\sigma}}_{12}(t) = & \hat{X}_{12}(t) - i \left[\omega_{21} - \frac{i}{2} A_2 \right] \hat{\sigma}_{12}(t) \\ & + (\Gamma_{2123} + \Gamma_{3212}^*) \hat{\sigma}_{23}(t), \end{aligned} \quad (3.21d)$$

$$\begin{aligned} \dot{\hat{\sigma}}_{23}(t) = & \hat{X}_{23}(t) - i \left[\omega_{32} - \frac{i}{2} A_3 - \frac{i}{2} A_2 \right] \hat{\sigma}_{23}(t) \\ & + (\Gamma_{1232} + \Gamma_{2321}^*) \hat{\sigma}_{12}(t), \end{aligned} \quad (3.21e)$$

$$\dot{\hat{\sigma}}_{13}(t) = \hat{X}_{13}(t) - i \left[\omega_{31} - \frac{i}{2} A_3 \right] \hat{\sigma}_{13}(t). \quad (3.21f)$$

Here $A_2 = 2\beta_{2112}$ and $A_3 = 2\beta_{3223}$ are the Einstein A coefficients for the $2 \rightarrow 1$ and $3 \rightarrow 2$ transitions, respectively, and

$$\Delta_{ji} = - \sum_m (\gamma_{jmmj} - \gamma_{immi}) \quad (3.22)$$

is the level shift (nonrelativistic and prior to mass renormalization) of the $j \rightarrow i$ transition. We have incorporated this shift into ω_{ji} , so that ω_{ji} refers to the shifted frequency. We have also defined the operators

$$\begin{aligned} \hat{X}_{ij}(t) = & \sum_{k,\lambda} \sum_{l,m} D_{k\lambda lm} \{ [\delta_{jl} \hat{\sigma}_{im}(t) - \delta_{im} \hat{\sigma}_{lj}(t)] \hat{a}_{k\lambda}(0) e^{-i\omega_k t} \\ & + \hat{a}_{k\lambda}^\dagger(0) e^{i\omega_k t} [\delta_{jl} \hat{\sigma}_{im}(t) - \delta_{im} \hat{\sigma}_{lj}(t)] \}. \end{aligned} \quad (3.23)$$

In the adiabatic approximation $\hat{\sigma}_{ij}$ oscillates approximately at the frequency ω_{ji} . We have therefore made a rotating-wave approximation in writing equations (3.21), ignoring terms on the right-hand side that do not oscillate near the principal frequency of the term on the left-hand side. Note that, unless ω_{21} and ω_{32} are close to each other, the last term on the right-hand side of both (3.21d) and (3.21e) may be dropped as a rotating-wave term. If ω_{21} and ω_{32} are approximately equal to each other, however, these terms must in general be retained. Terms of this type were obtained by Landau in 1927 and subsequently rediscovered by several workers.²²⁻²⁴ The renormalization of the generalized frequency-shift terms has been discussed by Cardimona and Stroud.²⁵

IV. INTENSITY CORRELATION IN TERMS OF ATOMIC OPERATORS

We recall from Eq. (2.4) that the normally ordered field correlation function $\Gamma^{(2,2)}(x_1; x_2)$ involves products of the vector potential operators $\hat{\mathbf{A}}^{(+)}$ and $\hat{\mathbf{A}}^{(-)}$ evaluated in the far field of the atomic radiation. These quantities are derived from the mode expansion for $\hat{\mathbf{A}}$, Eq. (3.5), together with Eq. (3.15a). To lowest order in $1/r$ and for $t > r/c$, we find that

$$\hat{\mathbf{A}}(\mathbf{r}, t) = \hat{\mathbf{A}}^{(+)}(\mathbf{r}, t) + \hat{\mathbf{A}}^{(-)}(\mathbf{r}, t), \quad (4.1)$$

where

$$\hat{\mathbf{A}}^{(+)}(\mathbf{r}, t) = \hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t) + \hat{\mathbf{A}}_S^{(+)}(\mathbf{r}, t), \quad (4.2)$$

$$\hat{\mathbf{A}}^{(-)}(\mathbf{r}, t) = [\hat{\mathbf{A}}^{(+)}(\mathbf{r}, t)]^\dagger,$$

$$\hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t) = \sum_{\mathbf{k}, \lambda} \left[\left[\frac{2\pi\hbar c^2}{\omega_k V} \right]^{1/2} \epsilon_{\mathbf{k}\lambda} \hat{a}_{\mathbf{k}\lambda}(0) e^{-i(\omega_k t - \mathbf{k} \cdot \mathbf{r})} \right], \quad (4.3)$$

$$\begin{aligned} \mathbf{A}_S^{(+)}(\mathbf{r}, t) = & -i\mathbf{h}_{12}(\mathbf{r})\hat{\sigma}_{12}(t-r/c) \\ & -i\mathbf{h}_{23}(\mathbf{r})\hat{\sigma}_{23}(t-r/c), \end{aligned} \quad (4.4)$$

$$\mathbf{h}_{lm}(\mathbf{r}) \equiv \left[\frac{\boldsymbol{\mu}_{lm}}{r} - \frac{(\boldsymbol{\mu}_{lm} \cdot \mathbf{r})\mathbf{r}}{r^3} \right] k_{lm}. \quad (4.5)$$

We have once again resorted to the adiabatic approximation,

$$\hat{\sigma}_{lm}(t+\tau) \cong \hat{\sigma}_{lm}(t) e^{-i\omega_{lm}\tau}, \quad (4.6)$$

and have defined $k_{lm} \equiv |\omega_{lm}|/c$. The vector-potential field operator is seen to be the sum of two terms—the free-field (vacuum) operator $\hat{\mathbf{A}}_F$ plus the source-field operator $\hat{\mathbf{A}}_S$ associated with the atomic radiation. $\hat{\mathbf{A}}$ has the same form that one would expect from classical considerations, if the association

$$\begin{aligned} \hat{\mathbf{D}}(t) \equiv & -\omega_{21}^2 \boldsymbol{\mu}_{12} [\hat{\sigma}_{12}(t) + \hat{\sigma}_{21}(t)] \\ & -\omega_{32}^2 \boldsymbol{\mu}_{23} [\hat{\sigma}_{23}(t) + \hat{\sigma}_{32}(t)] \end{aligned} \quad (4.7)$$

with $d^2\hat{\boldsymbol{\mu}}(t)/dt^2$ is made, with $\hat{\boldsymbol{\mu}}$ as the dipole moment operator. Note that in the decomposition of the vector potential operator into positive- and negative-frequency components as required in (2.4) and as made in (4.1), one must in general exercise care.²⁶ However, it is sufficient for our purposes to consider time intervals such that $\omega t \gg 1$, so that (4.1)–(4.4) follow without ambiguity.²⁷

$\Gamma^{(2,2)}(x_1; x_2)$ now follows by combining (4.1)–(4.5) with (2.4) for the initial state

$$|\Psi\rangle = |\psi_A\rangle \otimes | \{0\} \rangle. \quad (4.8)$$

Written explicitly for $t_2 \geq t_1$, the time-ordered correlation function becomes

$$\begin{aligned} \Gamma^{(2,2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = & \langle [\hat{\mathbf{A}}_F^{(-)}(\mathbf{r}_1, t_1) + i\mathbf{h}_{12i}(\mathbf{r}_1)\hat{\sigma}_{21}(t_1-r_1/c) + i\mathbf{h}_{23i}(\mathbf{r}_1)\hat{\sigma}_{32}(t_1-r_1/c)] \\ & \times [\hat{\mathbf{A}}_F^{(-)}(\mathbf{r}_2, t_2) + i\mathbf{h}_{12j}(\mathbf{r}_2)\hat{\sigma}_{21}(t_2-r_2/c) + i\mathbf{h}_{23j}(\mathbf{r}_2)\hat{\sigma}_{32}(t_2-r_2/c)] \\ & \times [\hat{\mathbf{A}}_F^{(+)}(\mathbf{r}_2, t_2) - i\mathbf{h}_{12j}(\mathbf{r}_2)\hat{\sigma}_{12}(t_2-r_2/c) - i\mathbf{h}_{23j}(\mathbf{r}_2)\hat{\sigma}_{23}(t_2-r_2/c)] \\ & \times [\hat{\mathbf{A}}_F^{(+)}(\mathbf{r}_1, t_1) - i\mathbf{h}_{12i}(\mathbf{r}_1)\hat{\sigma}_{12}(t_1-r_1/c) - i\mathbf{h}_{23i}(\mathbf{r}_1)\hat{\sigma}_{23}(t_1-r_1/c)] \rangle. \end{aligned} \quad (4.9)$$

To proceed further we require certain commutation relations between the free field and the atomic operators appearing in (4.9). Making use of the procedure discussed by Renaud *et al.*²⁸ generalized to the case of three levels,^{29(b)} we evaluate these commutators in the Appendix and find the following results, valid once again in the adiabatic approximation and in the far field.

$$\begin{aligned} \text{A. } t-r/c > t', \text{ with } t-r/c-t' & \gg \frac{1}{\omega} \\ [\hat{\sigma}_{12}(t'), \hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t)] & = 0 \text{ and H.c.}, \\ [\hat{\sigma}_{23}(t'), \hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t)] & = 0 \text{ and H.c.} \end{aligned} \quad (4.10)$$

$$\text{B. } t-r/c = t', \text{ with } t' \gg 1/\omega, t-r/c \gg 1/\omega$$

$$\begin{aligned} [\hat{\sigma}_{12}(t'), \hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t)] & = \frac{i}{2} \mathbf{h}_{23}(\mathbf{r}) \hat{\sigma}_{13}(t') \text{ and H.c.}, \\ [\hat{\sigma}_{23}(t'), \hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t)] & = -\frac{i}{2} \mathbf{h}_{12}(\mathbf{r}) \hat{\sigma}_{13}(t') \text{ and H.c.} \end{aligned} \quad (4.11)$$

These commutation relations are crucial to our treatment. Corresponding results have been obtained for the case of a two-level atom,^{27,28,30,31} but the restricted Hilbert space in that case produces no contributions analogous to those of Eq. (4.11). A quite general argument presented by Mol-

low³⁰ for evaluating commutation relations between free-field and atomic operators holds under the assumption of a localized (atomic) current distribution. While the current distribution appropriate to the Coulomb gauge is a transverse delta function, which is not local, a simple extension of Mollow's argument can nonetheless be used to evaluate certain atom-field commutators, namely, those involving the electric and magnetic field operators. However, we have relied upon the procedure outlined in the Appendix. Since the procedure involves the adiabatic approximation, Eqs. (4.10) and (4.11) are valid only on a time scale that is large compared to the optical period.

In evaluating (4.9) we will consider the two cases A and

B of (4.10) and (4.11) separately. To make explicit the time-ordering requirement associated with the detection process, we will continue to assume $t_2 \geq t_1$. The commutation relations (4.10) and (4.11) as applied to (4.9) in this case require that $t_2 - r_2/c \geq t_1 - r_1/c$. In other words, for the times specified above, $\Gamma^{(2,2)}$ is associated with a time ordering in the atomic frame of reference.

Case A: $t_2 - r_2/c > t_1 - r_1/c$. In this case terms involving the free-field operators $\hat{\mathbf{A}}_F^{(\pm)}$ make no contribution, since

$$\hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t) | \{0\} \rangle = 0 \text{ and H.c.} \quad (4.12)$$

Combining (3.9), (4.9), (4.10), and (4.12) we obtain

$$\begin{aligned} \Gamma^{(2,2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = & h_{12}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) \langle \hat{\sigma}_{21}(1) \hat{\sigma}_{22}(2) \hat{\sigma}_{12}(1) \rangle + h_{12j}(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) h_{23j}(\mathbf{r}_1) \langle \hat{\sigma}_{21}(1) \hat{\sigma}_{22}(2) \hat{\sigma}_{23}(1) \rangle \\ & + h_{12}^2(\mathbf{r}_1) h_{23}^2(\mathbf{r}_2) \langle \hat{\sigma}_{21}(1) \hat{\sigma}_{33}(2) \hat{\sigma}_{12}(1) \rangle + h_{12j}(\mathbf{r}_1) h_{23}^2(\mathbf{r}_2) h_{23j}(\mathbf{r}_1) \langle \hat{\sigma}_{21}(1) \hat{\sigma}_{33}(2) \hat{\sigma}_{23}(1) \rangle \\ & + h_{23j}(\mathbf{r}_1) h_{12j}(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) \langle \hat{\sigma}_{32}(1) \hat{\sigma}_{22}(2) \hat{\sigma}_{12}(1) \rangle + h_{23}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) \langle \hat{\sigma}_{32}(1) \hat{\sigma}_{22}(2) \hat{\sigma}_{23}(1) \rangle \\ & + h_{23j}(\mathbf{r}_1) h_{23}^2(\mathbf{r}_2) h_{12j}(\mathbf{r}_1) \langle \hat{\sigma}_{32}(1) \hat{\sigma}_{33}(2) \hat{\sigma}_{12}(1) \rangle + h_{23}^2(\mathbf{r}_1) h_{23}^2(\mathbf{r}_2) \langle \hat{\sigma}_{32}(1) \hat{\sigma}_{33}(2) \hat{\sigma}_{23}(1) \rangle, \end{aligned} \quad (4.13)$$

where (1) $\equiv (t_1 - r_1/c)$, (2) $\equiv (t_2 - r_2/c)$, with $t_1 > r_1/c$, $t_2 > r_2/c$, and where a summation over the Cartesian component j is implicit. The evaluation of $\Gamma^{(2,2)}$ is thus reduced to the evaluation of a set of trilinear atomic correlation functions.^{23,27}

Consider, for instance,

$$\langle \hat{\sigma}_{32}(1) \hat{\sigma}_{22}(2) \hat{\sigma}_{23}(1) \rangle = \left\langle \hat{\sigma}_{32} \left[t_1 - \frac{r_1}{c} \right] \hat{\sigma}_{22} \left[t_2 - \frac{r_2}{c} \right] \hat{\sigma}_{23} \left[t_1 - \frac{r_1}{c} \right] \right\rangle = \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{22}(t + \tau) \hat{\sigma}_{23}(t) \rangle,$$

where $t = t_1 - r_1/c$, $\tau = t_2 - t_1 - r_2/c + r_1/c > 0$. From Eq. (3.21) we obtain

$$\begin{aligned} \frac{\partial}{\partial \tau} \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{22}(t + \tau) \hat{\sigma}_{23}(t) \rangle = & -A_2 \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{22}(t + \tau) \hat{\sigma}_{23}(t) \rangle \\ & + A_3 \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{33}(t + \tau) \hat{\sigma}_{23}(t) \rangle \\ & + \langle \hat{\sigma}_{32}(t) \hat{X}_{22}(t + \tau) \hat{\sigma}_{23}(t) \rangle, \end{aligned} \quad (4.14a)$$

$$\begin{aligned} \frac{\partial}{\partial \tau} \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{33}(t + \tau) \hat{\sigma}_{23}(t) \rangle = & -A_3 \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{33}(t + \tau) \hat{\sigma}_{23}(t) \rangle \\ & + \langle \hat{\sigma}_{32}(t) \hat{X}_{33}(t + \tau) \hat{\sigma}_{23}(t) \rangle. \end{aligned} \quad (4.14b)$$

To proceed we once again require certain commutation relations between the free-field operators $\hat{\mathbf{A}}^{(\pm)}(0, t)$, which are now evaluated at the position of the atom instead of in the far field [(4.10) and (4.11)], and atomic operators $\hat{\sigma}_{lm}(t')$. The derivation is presented in the Appendix. We find from Eqs. (3.23), (A13), and (A14), for the state (4.8),

$$\begin{aligned} f(t, \tau) \equiv \langle \hat{\sigma}_{32}(t) \hat{X}_{22}(t + \tau) \hat{\sigma}_{23}(t) \rangle = & 0, \quad t > 0 \\ = \langle \hat{\sigma}_{33}(t) \rangle (A_2 + A_3), \quad \tau = 0 \end{aligned} \quad (4.15)$$

and

$$\langle \hat{\sigma}_{32}(t) \hat{X}_{33}(t + \tau) \hat{\sigma}_{23}(t) \rangle = 0, \quad \tau \geq 0. \quad (4.16)$$

With (4.16) the solution of (4.14b) is

$$\langle \hat{\sigma}_{32}(t) \hat{\sigma}_{33}(t + \tau) \hat{\sigma}_{23}(t) \rangle = \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{33}(t) \hat{\sigma}_{23}(t) \rangle e^{-A_3 \tau} = 0$$

because of (3.9). From (4.14a) and (4.15) we then have

$$\begin{aligned} \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{22}(t + \tau) \hat{\sigma}_{23}(t) \rangle = & \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{22}(t) \hat{\sigma}_{23}(t) \rangle e^{-A_2 \tau} \\ & + \int_0^\tau e^{A_2(\tau - \tau')} f(t, \tau') d\tau', \\ \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{22}(t + \tau) \hat{\sigma}_{23}(t) \rangle = & \langle \hat{\sigma}_{33}(t) \rangle e^{-A_2 \tau} + \int_0^\tau e^{A_2(\tau - \tau')} f(t, \tau') d\tau', \end{aligned}$$

where again we have used (3.9). From (3.21c) it follows that

$$\langle \hat{\sigma}_{33}(t) \rangle = \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3 t} \quad (4.17)$$

and therefore that

$$\begin{aligned} \langle \hat{\sigma}_{32}(t) \hat{\sigma}_{22}(t + \tau) \hat{\sigma}_{23}(t) \rangle = & \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3 t} e^{-A_2 \tau} \\ & + \int_0^\tau e^{A_2(\tau - \tau')} f(t, \tau') d\tau'. \end{aligned} \quad (4.18)$$

The integral term in this equation arises from the nonvan-

ishing commutators of atomic and free-field operators for equal-time arguments. As we have already pointed out in connection with Eqs. (4.10) and (4.11), we expect the function $f(t, \tau)$ to be nonzero only over a range in τ on the order of the optical period. Hence the integral term in (4.18) contributes a correction of order (Einstein A coefficient)/(optical period) and must be dropped to be consistent with our earlier approximations. Therefore,

$$\langle \hat{\sigma}_{32}(t) \hat{\sigma}_{22}(t+\tau) \hat{\sigma}_{23}(t) \rangle = \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3 t} e^{-A_2 \tau}, \quad \tau \geq 0. \quad (4.19)$$

By continuing this analysis, we can show that in fact $\langle \hat{\sigma}_{32}(1) \hat{\sigma}_{22}(2) \hat{\sigma}_{23}(1) \rangle$ is the only nonvanishing atomic correlation function appearing in (4.13). We thus find

$$\begin{aligned} \Gamma^{(2,2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) &= h_{23}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) \langle \hat{\sigma}_{33}(0) \rangle \exp[-A_3(t_1 - r_1/c)] \\ &\times \exp\{-A_2[(t_2 - t_1) - (r_2 - r_1)/c]\}, \end{aligned} \quad (4.20)$$

with $t_2 \geq t_1$, $t_2 - r_2/c > t_1 - r_1/c$. Of course for the initial state $|\psi_A\rangle = |3\rangle$, $\langle \hat{\sigma}_{33}(0) \rangle = 1$. Note that (4.5) allows the angular dependence of the dipole field to be expressed in the form

$$h_{23}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) = \frac{\omega_{32}^2 \mu_{23}^2 \sin^2 \theta_1}{c^2 r_1^2} \frac{\omega_{21}^2 \mu_{12}^2 \sin^2 \theta_2}{c^2 r_2^2}, \quad (4.21)$$

where θ_1 is the angle between \mathbf{r}_1 and $\boldsymbol{\mu}_{23}$ and θ_2 is the angle between \mathbf{r}_2 and $\boldsymbol{\mu}_{12}$.

Case B: $t_2 - r_2/c = t_1 - r_1/c \equiv T$. In this case terms involving the free-field operators $\hat{\mathbf{A}}_F^{(\pm)}$ do contribute to $\Gamma^{(2,2)}$. From (4.9), (4.11), and (4.12), we have

$$\begin{aligned} \Gamma^{(2,2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) &= \langle \{ -[h_{12i}(\mathbf{r}_1) \hat{\sigma}_{21}(T) + h_{23i}(\mathbf{r}_1) \hat{\sigma}_{32}(T)] [h_{12j}(\mathbf{r}_2) \hat{\sigma}_{21}(T) + h_{23j}(\mathbf{r}_2) \hat{\sigma}_{32}(T)] \\ &\quad - \frac{1}{2} h_{12i}(\mathbf{r}_1) h_{23j}(\mathbf{r}_2) \hat{\sigma}_{31}(T) + \frac{1}{2} h_{23i}(\mathbf{r}_1) h_{12j}(\mathbf{r}_2) \hat{\sigma}_{31}(T) \} \\ &\quad \times \{ -[h_{12j}(\mathbf{r}_2) \hat{\sigma}_{12}(T) + h_{23j}(\mathbf{r}_2) \hat{\sigma}_{23}(T)] [h_{12i}(\mathbf{r}_1) \hat{\sigma}_{12}(T) + h_{23i}(\mathbf{r}_1) \hat{\sigma}_{23}(T)] \\ &\quad - \frac{1}{2} h_{12i}(\mathbf{r}_1) h_{23j}(\mathbf{r}_2) \hat{\sigma}_{13}(T) + \frac{1}{2} h_{23i}(\mathbf{r}_1) h_{12j}(\mathbf{r}_2) \hat{\sigma}_{13}(T) \} \rangle. \end{aligned} \quad (4.22)$$

In this equation terms in $\hat{\sigma}_{13}$ and $\hat{\sigma}_{31}$ arise from the nonzero commutators (4.11). Carrying out the multiplication in (4.22), we find with the help of (3.9) and (4.17)

$$\begin{aligned} \Gamma^{(2,2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) &= \frac{1}{4} [h_{23i}(\mathbf{r}_1) h_{12j}(\mathbf{r}_2) + h_{23j}(\mathbf{r}_2) h_{12i}(\mathbf{r}_1)]^2 \\ &\times \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t_1 - r_1/c)} \end{aligned} \quad (4.23)$$

with $t_2 - r_2/c = t_1 - r_1/c$ and $t_2 \geq t_1$.

Examination of our results in cases A and B leads to the following conclusions. When $t_2 - r_2/c > t_1 - r_1/c$, the only contribution to $\Gamma^{(2,2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2)$ is seen from (4.20) to be the process

$$\omega_{32} \rightarrow \mathbf{r}_1, \quad (4.24a)$$

$$\omega_{21} \rightarrow \mathbf{r}_2$$

in which a photon is detected at \mathbf{r}_1 from the transition $3 \rightarrow 2$, and then a photon is detected at \mathbf{r}_2 from the $2 \rightarrow 1$ transition. When $t_2 - r_2/c = t_1 - r_1/c$, however, we also expect a contribution to $\Gamma^{(2,2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2)$ from the process

$$\omega_{32} \rightarrow \mathbf{r}_2, \quad (4.24b)$$

$$\omega_{21} \rightarrow \mathbf{r}_1$$

and indeed (4.23) exhibits contributions from both processes (4.24a) and (4.24b). The first term in square brackets in (4.23) is proportional to the amplitude for finding a

photon from the $3 \rightarrow 2$ transition at the position \mathbf{r}_1 and a photon from the $2 \rightarrow 1$ transition at the position \mathbf{r}_2 . Likewise, the second term is proportional to the amplitude for a $3 \rightarrow 2$ photon at \mathbf{r}_2 and a $2 \rightarrow 1$ photon at \mathbf{r}_1 . If the points \mathbf{r}_1 and \mathbf{r}_2 coincide, the possibility of different paths to the detectors is eliminated in the experiment envisioned, even if atomic recoil associated with the emission process is taken into account. For zero time delay, it is impossible to distinguish between the processes (4.24a) and (4.24b). Thus we can interpret (4.23) as representing the intrinsic indistinguishability of these two decay channels: probability amplitudes are added, then squared.

It is worth emphasizing that our results are based on the adiabatic (i.e., Weisskopf-Wigner) and rotating-wave approximations. However, we do expect that the interference between the two decay channels will persist, at most, only over a time on the order of an optical period. Thus the abrupt cutoff of the interference for nonzero time delay, as derived above within our approximations, will be quite accurate for our purposes.

V. JOINT PROBABILITY OF PHOTOELECTRIC DETECTION

Given the expressions of Sec. IV for the fourth-order field correlation functions $\Gamma^{(2,2)}$, we can now address the question of actual photoelectric detection probabilities. We return to Eq. (2.3) written explicitly to reflect the time ordering associated with the detection process:

$$P_2(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = k_0^2 \int_{t_1}^{t_1 + \Delta t} dt'_1 \int_{t_2}^{t_2 + \Delta t} dt'_2 [U(t'_2 - t'_1) \Gamma^{(2,2)}(\mathbf{r}_1, t'_1; \mathbf{r}_2, t'_2) + U(t'_1 - t'_2) \Gamma^{(2,2)}(\mathbf{r}_1, t'_1; \mathbf{r}_2, t'_2)], \quad (5.1)$$

where we have taken the resolution intervals $\Delta t_1 = \Delta t_2 = \Delta t$ and where $U(t)$ is the unit step function. For definiteness we will consider the case $t_2 \geq t_1$ and will furthermore assume that $|\mathbf{r}_1| = |\mathbf{r}_2|$ in order to avoid a rather cumbersome notation that accounts for retardation. The expression for $\Gamma^{(2,2)}$ for $t'_2 \geq t'_1$ has been obtained in Sec. IV, Eqs. (4.20) and (4.23). A corresponding result for $\Gamma^{(2,2)}$ for $t'_1 \geq t'_2$ follows from Eq. (4.9) with the change $(\mathbf{r}_1, t_1) \rightarrow (\mathbf{r}_2, t'_2)$ and $(\mathbf{r}_2, t_2) \rightarrow (\mathbf{r}_1, t'_1)$. Substituting these expressions for $\Gamma^{(2,2)}$ into (5.1), we find

$$P_2(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = k_0^2 \int_{t_1}^{t_1 + \Delta t} dt'_1 \int_{t_2}^{t_2 + \Delta t} dt'_2 [U(t'_2 - t'_1) h_{23}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t'_1 - r_1/c)} e^{-A_2(t'_2 - t'_1)} + U(t'_1 - t'_2) h_{23}^2(\mathbf{r}_2) h_{12}^2(\mathbf{r}_1) \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t'_2 - r_2/c)} e^{-A_2(t'_1 - t'_2)}] \quad (5.2)$$

with $t_2 \geq t_1$ and $|\mathbf{r}_1| = |\mathbf{r}_2|$. The terms that arise from the nonzero commutation relation (4.11) and that were discussed in connection with Eq. (4.23) can be shown to make a contribution at most of order $1/\omega\Delta t$. Since in practice the detection interval $\Delta t \geq 10^{-10}$ sec, these terms have been dropped in writing (5.2) consistent with the rotating-wave approximation in (2.1). While the detection interval is long compared to the optical period, it can however be quite short as compared to the atomic lifetime. In this case we set $t'_1 = t_1$ and $t'_2 = t_2$ in the exponential factors and evaluate the remaining integrals, with the result

$$P_2(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = k_0^2 \langle \hat{\sigma}_{33}(0) \rangle \left\{ h_{23}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) e^{-A_3(t_1 - r_1/c)} e^{-A_2(t_2 - t_1)} \times \left[U(t_2 - t_1 - \Delta t) (\Delta t)^2 + U(t_1 + \Delta t - t_2) \left(\frac{(t_2 - t_1 + \Delta t)^2}{2} - (t_2 - t_1)^2 \right) \right] + h_{23}^2(\mathbf{r}_2) h_{12}^2(\mathbf{r}_1) e^{-A_3(t_2 - r_2/c)} e^{-A_2(t_1 - t_2)} U(t_1 + \Delta t - t_2) \frac{(t_2 - t_1 - \Delta t)^2}{2} \right\}, \quad (5.3)$$

$t_2 \geq t_1$, $|\mathbf{r}_1| = |\mathbf{r}_2|$. Examination of two special cases leads to a straightforward interpretation of this equation.

A. $t_2 \geq t_1 + \Delta t$:

$$P_2(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = k_0^2 \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t_1 - r_1/c)} \times e^{-A_2(t_2 - t_1)} h_{23}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) (\Delta t)^2. \quad (5.4)$$

B. $t_1 = t_2$:

$$P_2(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = k_0^2 \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t_1 - r_1/c)} \frac{(\Delta t)^2}{2} \times [h_{23}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) + h_{23}^2(\mathbf{r}_2) h_{12}^2(\mathbf{r}_1)]. \quad (5.5)$$

Equation (5.4) corresponds to the detection probability that would be obtained from our earlier result (4.20) and (2.3) in the absence of any time ordering of the field correlation function $\Gamma^{(2,2)}$. Equation (5.5) is analogous to our result (4.23) in that it allows for the possibility of detection via either of the two processes indicated in (4.24). We may ask why (5.5) involves the sum of probabilities for the possible detection channels (4.24), while $\Gamma^{(2,2)}$ in (4.23) involves a sum of probability amplitudes, which is then squared. The answer is of course that for times short compared to $1/\omega_{21}$ or $1/\omega_{32}$ it is in principle impossible to distinguish between the decay channels. For detection times long compared to $1/\omega$, this intrinsic indistinguishability is still present but contributes only to order $1/\omega\Delta t$ as compared to unity. Over long detection intervals we are still unable to resolve the two decay channels of (4.24), but now the indistinguishability arises from our lack of resolution in the apparatus and not from a fundamental

inability to separate the two possibilities. Over most of the interval Δt , the two decay channels are in principle distinguishable, and therefore the probabilities add as in (5.3). The factors in $(t_1, t_2, \Delta t)$ in (5.3) show that as $t_2 - t_1$ increases from zero to Δt , the two decay channels become increasingly resolvable and over a decreasing fraction of the interval Δt is it likely to have recorded the ω_{21} photon first.

Note that without the time ordering in the original expression (2.1) for the joint probability of photoelectric detection, we would not have obtained the expressions shown in (5.3)–(5.5), but would have instead found (5.4) over the entire range $t_2 \geq t_1$. The time ordering of (2.1) is essential if our analysis is to produce physically reasonable results. Although the standard derivation of (2.1) leads to the time-ordered field correlation function, quite often one deals with fields that may be taken as effectively free, for which the time ordering is not important.

If we now return to Eq. (5.3) and consider the case of a single photoelectric detector at $\mathbf{r}_1 = \mathbf{r}_2 = \mathbf{r}$, we are able to express the joint probability of photodetection in a particularly simple form as a product of two factors,

$$P_2(\mathbf{r}, t_1; \mathbf{r}, t_2) = P_1^{(3,2)}(\mathbf{r}, t_1) P_1^{(2,1)}(\mathbf{r}, t_2 - t_1), \quad (5.6)$$

where

$$P_1^{(3,2)}(\mathbf{r}, t) = k_0 h_{23}^2(\mathbf{r}) \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t - r/c)} \Delta t, \quad (5.7)$$

and

$$P_1^{(2,1)}(\mathbf{r}, t) = k_0 h_{12}^2(\mathbf{r}) e^{-A_2(t - r/c)} \Delta t. \quad (5.8)$$

As before, we assume that Δt is much shorter than either of the atomic lifetimes and take $t_2 \geq t_1$.

The two terms $P_1^{(i,j)}(\mathbf{r}, t)$ are probability densities for

single photoelectric detection events as would be obtained from Eq. (2.5). $P_1^{(3,2)}(\mathbf{r}, t)$ is the probability for detection of a photon of approximate frequency ω_{32} from the transition $3 \rightarrow 2$ for an atom initially excited to the state $|3\rangle$ with probability $\langle \hat{\sigma}_{33}(0) \rangle$. $P_1^{(2,1)}(\mathbf{r}, t)$ likewise gives the probability for detection of a photon of approximate frequency ω_{21} from the transition $2 \rightarrow 1$ for an atom excited to the state $|2\rangle$ with unit probability. Equation (5.6) thus suggests an interpretation of the atomic cascade as arising from sequential but otherwise statistically independent events. The first photoelectric detection at (\mathbf{r}_1, t_1) ensures that the atom is in the state $|2\rangle$ at time $(t_1 - r_1/c)$, from which it decays over a lifetime $(A_2)^{-1}$ in a fashion independent of the time taken to initially populate $|2\rangle$. As was found for resonance fluorescence from a two-level atom,^{9,27,32,33,29(a)} we likewise find that the atomic cascade proceeds in discrete "quantum" jumps. The only exception to this statement is for zero time delay between the two events. In this case the cascade cannot be viewed as a sequential process but rather as a superposition of the two processes as indicated by (4.24). Any manifestation of this interference between decay channels is however lost in the act of detection because of the condition $\omega\Delta t \gg 1$.

Equation (5.3) can be cast into a form which avoids such explicit reference to the properties of the detector arrangement by imagining the atom to be surrounded by two closely spaced spherical detectors, again in the far field. The joint probability $P_2(t, t + \tau)$ for photoelectric detection at times t and $t + \tau$ is given in this case from Eq. (5.3) by introducing the definition

$$k_0 = \kappa_0 dS, \quad (5.9)$$

with dS as the differential element of surface area of one of the detectors, and by summing the resultant expression for $P_2(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2)$ over all elements of the detectors. We find for $\Delta t \ll A_2^{-1}, A_3^{-1}$,

$$P_2(t, t + \tau) = \frac{4\pi^2 \hbar^2 c^2 \kappa_0^2}{\omega_{32} \omega_{21}} [A_3 \Delta t \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t-r/c)}] \times (A_2 \Delta t e^{-A_2 \tau}), \quad t > r/c, \tau \geq 0 \quad (5.10)$$

where r is the distance from the atom to the detectors and where Eq. (3.19) has been used to eliminate μ_{lm}^2 . The fraction in (5.10) expresses the effective efficiency of the

detectors. For the ideal detector considered, we will set this term to unity, yielding

$$P_2(t, t + \tau) = [A_3 \Delta t \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t-r/c)}] (A_2 \Delta t e^{-A_2 \tau}), \quad t > r/c, \tau \geq 0. \quad (5.11)$$

Note that the sum over initial detection times t and delays τ of the joint probability $P_2(t, t + \tau)$ gives the likelihood P_2 of two sequential photoelectric events over the entire course of the cascade. From (5.11) we obtain

$$P_2 = \int_{r/c}^{\infty} dt \int_0^{\infty} d\tau P_2(t, t + \tau) = \langle \hat{\sigma}_{33}(0) \rangle. \quad (5.12)$$

Although this result might have been anticipated, it is nonetheless remarkable in its simplicity. Equations (5.6), (5.11), and (5.12) demonstrate that initial atomic excitations involving a distribution of population among the levels (1,2,3), or coherences between these levels, do not affect the manner in which photon emissions occur, namely in a sequence of discrete quantum steps.

To conclude this section, we wish to consider a slightly different experimental arrangement than that depicted in Fig. 1. In measurements of the joint probability of photoelectric detection for an atomic cascade, frequency-selective filters are often employed in front of the photodetectors to unambiguously identify the atomic transition associated with the detected photon. In Fig. 1(b) assume that narrow bandpass filters centered at ω_{32} and ω_{21} are placed at \mathbf{r}_1 and \mathbf{r}_2 , respectively, and that ω_{32} and ω_{21} differ by an amount which is itself comparable to the transition frequencies. The photoelectric detection probabilities can be calculated in this case by returning to Eqs. (4.4) and (4.9). At the point \mathbf{r}_1 we include in $\hat{\mathbf{A}}^{(+)}(\mathbf{r}, t)$ only the contribution from the source field associated with the frequency ω_{32} , namely the $\hat{\sigma}_{23}$ term. Likewise at \mathbf{r}_2 we retain only the contribution from the source field of frequency ω_{21} , namely the $\hat{\sigma}_{12}$ term. This procedure corresponds to an admittedly naive picture of the frequency filter, yet it shall suffice for our purposes since we restrict ourselves to a time scale large compared to the inverse of the filter's bandwidth Γ . Note however that Γ can be chosen much greater than A_2 or A_3 so that we are still able to follow the evolution of decay.³⁴

Retaining only those terms in $\hat{\mathbf{A}}^{(\pm)}$ as discussed above, we proceed as in Secs. IV and V to arrive at the following expression for the joint probability of photoelectric detection $R_2(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2)$:

$$R_2(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = k_0^2 h_{23}^2(\mathbf{r}_1) h_{12}^2(\mathbf{r}_2) \langle \hat{\sigma}_{33}(0) \rangle e^{-A_3(t_1 - r_1/c)} e^{-A_2(t_2 - t_1)} \begin{cases} \Delta t^2, & t_1 + \Delta t \leq t_2 \\ -\frac{1}{2}(t_2 - t_1)^2 + \Delta t \left[t_2 - t_1 + \frac{\Delta t}{2} \right], & t_1 \leq t_2 \leq t_1 + \Delta t \\ \frac{1}{2}(t_2 - t_1)^2 + \Delta t \left[t_2 - t_1 + \frac{\Delta t}{2} \right], & t_1 - \Delta t \leq t_2 \leq t_1 \\ 0, & t_2 \leq t_1 - \Delta t \end{cases} \quad (5.13)$$

with $|\mathbf{r}_1| = |\mathbf{r}_2|$, $\Delta t \ll 1/A_2, 1/A_3$. Once again terms involving the commutators (4.10) and (4.11) have been dropped since they will contribute only to order $1/\omega\Delta t \ll 1$. The nonzero value of R_2 for $t_1 - \Delta t \leq t_2 \leq t_1$ is an obvious consequence of the finite resolution of the detectors. The term in brackets in (5.13) has a value of $(\Delta t)^2/2$ for $t_2 = t_1$ and rises to a value of $(\Delta t)^2$ for $t_2 \geq t_1 + \Delta t$. Relative to our result (5.3), there is thus a suppression of the zero-delay coincidence rate when frequency filters are used, corresponding to the loss of one of the two possible detection channels given by (5.5). The form given by (5.13) agrees well with that found in high-resolution measurements of atomic cascades.^{5,8}

VI. SUMMARY

Starting from the Hamiltonian (3.1)–(3.4), we have examined the question of photon correlations for a three-level atomic cascade. The dipole, rotating-wave, and adiabatic approximations have been employed in the calculation, and thus we expect that possible corrections to our results would be of the order of (Einstein A coefficient)/(transition frequency). Our analysis includes the possibility of a ladder scheme in which the transition frequencies are equal and for which Landau's generalized decay and frequency-shift terms must be considered. In agreement with the numerous observations quoted in the Introduction we find that, so far as photoelectric correlations are concerned, the atomic cascade proceeds in a fashion depending only upon the degree of excitation of the uppermost level. For nonzero time delay (intervals $\gg 1/\omega$) the emitted photons are statistically dependent in that the first photoelectric detection provides an unambiguous initiation for the second decay. The emissions are otherwise uncorrelated. For zero time delay between emissions there is an uncertainty as to which photon was emitted first [(4.20) and (4.23)] and an interference between possible decay channels. However this effect persists only over a very short interval, at most of order $1/\omega$, and hence is beyond the resolution of current experiments. Indeed, since the rotating-wave approximation is made consistently throughout our analysis, we are able to make only a qualitative assessment of the interference term. There is of course no contradiction between the absence of residual *temporal* correlation and the presence of polarization correlation, which arises from the symmetries of the initial and final states. Our calculation relates to the dynamical nature of the cascade.

As a final note, we return to the Weisskopf-Wigner result for the joint spectral density. In the frequency domain there is a definite correlation between emitted photons to ensure energy conservation in each microscopic event. The absence of such coupling in the time domain [(5.6) and (5.11)] results from the loss of frequency and hence energy information implicit in the measure-

ment of the temporal correlation of photons on a time scale much shorter than that of the lifetimes of the levels. Indeed, if one considers the circumstance described in Sec. V in which frequency-selective filters of bandpass Γ are placed between the photodetectors and the decaying atom, and if Γ is allowed to become comparable to A_2 or A_3 , the apparent independence of emitted photons in time is lost as the temporal and energy signatures of the process become mixed. An interpretation of the atomic cascade cannot be made without reference to the act of observation.

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APPENDIX

We require the following commutators:

$$[\hat{\sigma}_{lm}(t'), \hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t)] \text{ and H.c.} \quad (\text{A1})$$

$$[\hat{\sigma}_{lm}(t'), \hat{\mathbf{A}}_F^{(+)}(0, t)] \text{ and H.c.}, \quad (\text{A2})$$

with $l < m$. The position \mathbf{r} in (A1) is in the far field of the radiation from the atom at the origin. Our analysis is closely related to that of Renaud *et al.*²⁸

Consider first equation (A1),

$$\begin{aligned} \hat{\mathbf{C}} &\equiv [\hat{\sigma}_{lm}(t'), \hat{\mathbf{A}}_F^{(+)}(\mathbf{r}, t)] \\ &= \left[\hat{\sigma}_{lm}(t'), \sum_{\mathbf{k}, \lambda} \left[\frac{2\pi\hbar c^2}{\omega_k V} \right]^{1/2} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)} \boldsymbol{\epsilon}_{k\lambda} \hat{a}_{k\lambda}(0) \right], \end{aligned} \quad (\text{A3})$$

where the mode expression (3.5) has been used together with the free-field evolution for $\hat{a}_{k\lambda}(t)$. The operator $\hat{a}_{k\lambda}(0)$ may be eliminated from (A3) by using (3.15a) and the fact that *equal-time* atomic and field operators commute:

$$\begin{aligned} \hat{\mathbf{C}} &= - \sum_{\mathbf{k}, \lambda} \sum_{l', m'} \frac{2\pi c}{\omega_k V} \boldsymbol{\epsilon}_{k\lambda} (\boldsymbol{\mu}_{l'm'} \cdot \boldsymbol{\epsilon}_{k\lambda}) \omega_{m'l'} e^{i\mathbf{k} \cdot \mathbf{r}} e^{-i\omega_k(t-t')} \\ &\quad \times \left[\hat{\sigma}_{lm}(t'), \int_0^{t'} dt_1 e^{i\omega_k(t_1-t')} \hat{\sigma}_{l'm'}(t_1) \right]. \end{aligned} \quad (\text{A4})$$

Passing to the mode-continuum limit and using

$$\sum_{\lambda} \int d\Omega_k e^{i\mathbf{k} \cdot \mathbf{r}} \boldsymbol{\epsilon}_{k\lambda} (\boldsymbol{\mu}_{lm} \cdot \boldsymbol{\epsilon}_{k\lambda}) = 4\pi \frac{\sin(kr)}{kk_{lm}} \mathbf{h}_{lm}(\mathbf{r}), \quad (\text{A5})$$

where the integral is over solid angles about \mathbf{k} , we have

$$\hat{\mathbf{C}} = - \frac{1}{\pi} \sum_{l', m'} h_{l'm'}(\mathbf{r}) \left[\hat{\sigma}_{lm}(t'), \int_0^{t'} dt_1 \hat{\sigma}_{l'm'}(t_1) \int_0^{\infty} d\omega e^{-i\omega(t-t_1)} \sin \frac{\omega r}{c} \right]. \quad (\text{A6})$$

Thus far no approximations have been made. We show below that, within the adiabatic approximation, $\hat{\mathbf{C}} = 0$ for $l' > m'$, and furthermore that the integration over ω in (A6) may be extended to $-\infty$:

$$\hat{C} \cong -\frac{1}{\pi} \sum_{l' < m'} h_{l'm'}(\mathbf{r}) \left[\hat{\sigma}_{lm}(t'), \int_0^{t'} dt_1 \hat{\sigma}_{l'm'}(t_1) \int_{-\infty}^{\infty} d\omega e^{-i\omega(t-t_1)} \sin \frac{\omega r}{c} \right]. \quad (\text{A7})$$

Now

$$\begin{aligned} \int_0^{t'} dt_1 \hat{\sigma}_{l'm'}(t_1) \int_{-\infty}^{\infty} d\omega e^{-i\omega(t-t_1)} \sin \frac{\omega r}{c} \\ = -i\pi \int_0^{t'} dt_1 \hat{\sigma}_{l'm'}(t_1) [\delta(t-t_1-r/c) \\ - \delta(t-t_1+r/c)]. \quad (\text{A8}) \end{aligned}$$

For $t-r/c > t'$, therefore,

$$\int_0^{t'} dt_1 \hat{\sigma}_{l'm'}(t_1) \int_{-\infty}^{\infty} d\omega e^{-i\omega(t-t_1)} \sin \frac{\omega r}{c} = 0, \quad (\text{A9a})$$

whereas for $t-r/c = t'$ we have

$$\begin{aligned} \int_0^{t'} dt_1 \hat{\sigma}_{l'm'}(t_1) \int_{-\infty}^{\infty} d\omega e^{-i\omega(t-t_1)} \sin \frac{\omega r}{c} \\ = -\frac{i\pi}{2} \hat{\sigma}_{l'm'}(t'). \quad (\text{A9b}) \end{aligned}$$

Equations (A7) and (A9), together with the equal-time commutator (3.8), then give the commutator (A1) as written in (4.10) and (4.11).

It remains to justify the approximation (A7). This approximation of extending a frequency integration to $-\infty$

appears frequently in radiation theory. For instance, it was used by Fermi³⁵ in 1932, and in slightly different ways by Louisell³⁶ and Milonni and Knight.³⁷ Using Laplace transforms we can approach the problem in much the same way as these authors. Here we instead evaluate \hat{C} by making use of the adiabatic approximation, in which we are free to replace $\hat{\sigma}_{l'm'}(t_1)$ by $\sigma_{l'm'}(t') \exp[-i\omega_{m'l'}(t_1-t')]$. Making this approximation in (A6), we have

$$\begin{aligned} \hat{C} \cong -\frac{1}{\pi} \sum_{l', m'} h_{l'm'}(\mathbf{r}) [\hat{\sigma}_{lm}(t'), \hat{\sigma}_{l'm'}(t')] \\ \times \int_0^{\infty} d\omega \sin \left[\frac{\omega r}{c} \right] e^{-i\omega(t-t')} \\ \times \int_0^{t'} dt_1 \exp[i(\omega - \omega_{m'l'})(t_1-t')]. \quad (\text{A10}) \end{aligned}$$

For times long compared with an optical period the last integral is negligible when $\omega_{m'l'} < 0$. It is also clear that we introduce only a negligible error by letting ω take on all negative values when $\omega_{m'l'} > 0$. Thus

$$\begin{aligned} \hat{C} \cong -\frac{1}{\pi} \sum_{\substack{l', m' \\ l' < m'}} h_{l'm'}(\mathbf{r}) \left[\hat{\sigma}_{lm}(t'), \int_0^{t'} dt_1 \hat{\sigma}_{l'm'}(t_1) \int_{-\infty}^{\infty} d\omega \sin \left[\frac{\omega r}{c} \right] e^{-i\omega(t-t')} \exp[i(\omega - \omega_{m'l'})(t_1-t')] \right] \\ \cong -\frac{1}{\pi} \sum_{\substack{l', m' \\ l' < m'}} h_{l'm'}(\mathbf{r}) \left[\hat{\sigma}_{lm}(t'), \int_0^{t'} dt_1 \hat{\sigma}_{l'm'}(t_1) \int_{-\infty}^{\infty} d\omega \sin \left[\frac{\omega r}{c} \right] e^{-i\omega(t-t_1)} \right], \quad (\text{A11}) \end{aligned}$$

where in the last step we have again introduced the adiabatic approximation. The last expression is identical to (A7), which demonstrates the validity of the commutation relations (4.10) and (4.11) within the adiabatic approximation. In fact, precisely the same sort of approximation, extending an integration over ω to $-\infty$, may be used in the derivation of (4.4).

The derivation of the commutator (A2) proceeds similarly. The polarization summation and angular integration in (A4) now yield, for $\mathbf{r}=0$,

$$\sum_{\lambda} \int d\Omega_{\mathbf{k}} \epsilon_{\mathbf{k}\lambda} (\boldsymbol{\mu}_{lm} \cdot \boldsymbol{\epsilon}_{\mathbf{k}\lambda}) = \frac{8\pi}{3} \boldsymbol{\mu}_{lm}, \quad (\text{A12})$$

which when combined with an analysis similar to that above gives

$$[\hat{\sigma}_{lm}(t'), \hat{\mathbf{A}}_F^{(+)}(0, t)] \cong 0, \quad t > t', \quad l < m \quad (\text{A13})$$

$$\begin{aligned} [\hat{\sigma}_{lm}(t'), \hat{\mathbf{A}}_F^{(+)}(0, t)] \\ \cong -\sum_{m', l'} \frac{2\omega_{l'm'}}{3\pi c^2} \boldsymbol{\mu}_{l'm'} [\delta_{ml'} \hat{\sigma}_{lm'}(t') - \delta_{lm'} \hat{\sigma}_{l'm'}(t')] \\ \times (\pi\omega_{m'l'} - i\phi_{m'l'}), \quad t = t' \gg 1/\omega, \quad l < m \quad (\text{A14}) \end{aligned}$$

$$\phi_{ml} \equiv P \int_0^{\infty} \frac{\omega d\omega}{\omega - \omega_{ml}}. \quad (\text{A15})$$

Regarding (A14), it may be noted that the extension of the integration in ϕ_{ml} to $-\infty$ is *not* a good approximation at $\mathbf{r}=0$.³⁷

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